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Vanessa Preisler, Thomas Grange, Robson Ferreira, Louis-Anne de Vaulchier, Yves Guldner, Fransisco José Teran, Marek Potemski, Aristide Lemaître

Evidence for Excitonic Polarons in InAs/GaAs Quantum Dots

V. Preisler[*1], **T. Grange**[1], **R. Ferreira**[1], **L.A. de Vaulchier**[1], **Y. Guldner**[1], **F.J. Teran**[2], **M. Potemski**[2], and **A. Lemaître**[3]

[1] Laboratoire Pierre Aigrain, Ecole Normale Supérieure, 24 rue Lhomond, 75231 Paris Cedex 05, France
[2] Grenoble High Magnetic Field Laboratory, CNRS/MPI, 25 avenue des Martyrs, 38042 Grenoble Cedex 9, France
[3] Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France

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We investigate the interband transitions in several ensembles of self-assembled InAs/GaAs quantum dots by using photoluminescence excitation spectroscopy under strong magnetic field. Well defined resonances are observed in the spectra. A strong anticrossing between two transitions is observed in all samples, which cannot be accounted for by a purely excitonic model. The coupling between the mixed exciton-LO phonon states is calculated using the Fröhlich Hamiltonian. The excitonic polaron energies as well as the oscillator strengths of the interband transitions are determined. An anticrossing is predicted when two exciton-LO phonon states have close enough energies with phonon occupations which differ by one. A good agreement is found between the calculations and the experimental data evidencing the existence of excitonic polarons.

1 Introduction Various experimental and theoretical works have demonstrated that carriers confined in semiconductor quantum dots (QDs) are strongly coupled to the longitudinal optical (LO) vibrations of the underlying semiconductor lattice [1, 2, 3, 4, 5, 6, 7]. Far-infrared (FIR) magneto-absorption in n- and p-doped QDs have shown that intraband optical transitions involve polaron levels instead of the purely electronic states [1, 4, 8]. Recent theoretical works have shown that excitons in QDs strongly couple to LO phonons in spite of their electrical neutrality [9]. The eigenstates of the interacting exciton and phonon systems are the so-called excitonic polarons which are predicted to give significant modifications of the energy levels and large anticrossings when two exciton-phonon states have close enough energies with phonon occupations which differ by one. In the present work, we have studied, at T= 4 K, the photoluminescence excitation (PLE) under strong magnetic field up to 28 T of several ensembles of self-assembled InAs/GaAs QDs. PLE spectroscopy probes the absorption of a subensemble of similar QDs defined by the detection energy E_{det} and thus allows to circumvent part of the inhomogeneous broadening caused by dot size inhomogenity. Several well defined resonances are observed in all samples. The magnetic field dependence of the resonance energies allows an unambiguous assignment of the interband transitions. A strong anticrossing between two transitions is observed in all samples as the magnetic field is changed. Such an anticrossing cannot be accounted for by a purely excitonic model and one has to consider the exciton-lattice interactions. We have calculated the coupling between the mixed exciton-LO phonon states using the Fröhlich Hamiltonian and we have determined the excitonic polaron states as well as the energies and oscillator strengths of the interband transitions.

2 PLE Results Figure 1 depicts the PLE spectra of the three samples A1, A2 and A3 which were respectively undoped, n-doped and p-doped. The peaks observed are associated with transitions between

* Corresponding author: e-mail: vanessa.preisler@lpa.ens.fr

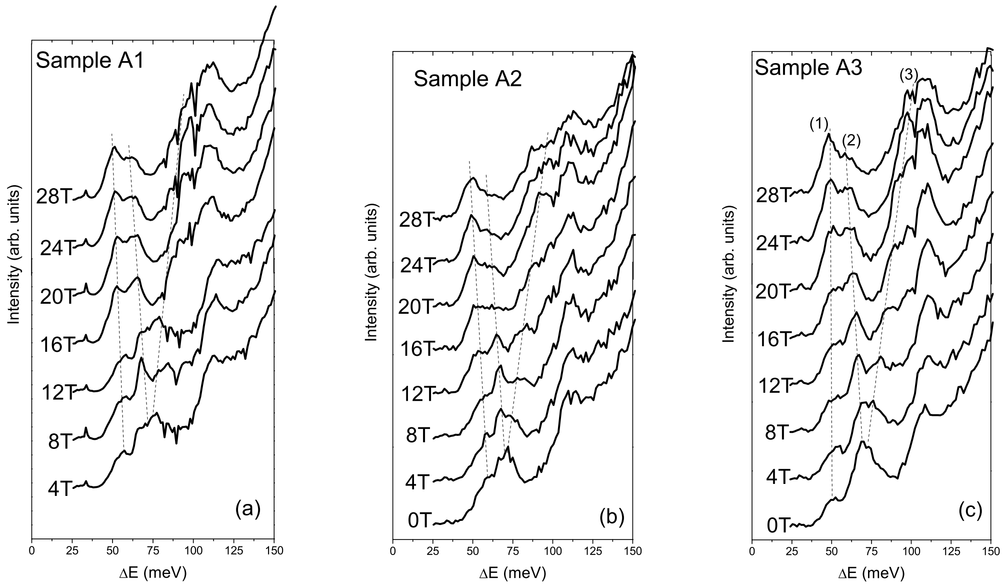

Fig. 1 Magneto-PLE spectra of samples A1 (a), A2 (b) and A3 (c) at 4 K from B=0 to B=28 T every 4 T and for a E_{det}=1215 meV. The dashed lines are guides for the eyes. Traces have been vertically offset for clarity.

bound levels in the QDs: a low-energy peak is observed at $\sim$50 meV, a second peak is found at $\sim$75 meV, a third peak is found at $\sim$110 meV. For QDs with a perfect cylindrical symmetry, the ground and first excited states in both the conduction and valence band are s and p-like respectively. In order to associate the excitation peaks with transitions in the QDs, a magnetic field B is applied along the sample's growth axis, as it is well known that the effect of a magnetic field is different for s, p and d states. The energy of the PLE peaks as a function of magnetic field is plotted in Fig. 2 for sample A3. As the magnetic field increases, the peak that was initially at $\sim$ 75 meV splits into two separate peaks: one peak that increases in energy and a second peak that decreases in energy. These peaks can be associated with p-like transitions. Let us now take a closer look at the oscillator strengths of the lower energy peaks, as shown in Fig. 1. At 0 T, the intensity of the low-energy peak at 50 meV is about 20% of that of the 75 meV peak. As the magnetic field increases, an exchange of oscillator strength between these two peaks is observed. For samples A1 and A3, at 20 T the two peaks have the same intensity and by 24 T the oscillator strength of the low-energy peak has surpassed that of its neighbor. For sample A2, the energy difference between the two peaks at 0T is smaller and therefore the anticrossing is observed at a lower magnetic field. Such a behavior can not be explained using a purely electronic model. It is necessary to use a model that takes into account the coupling between the optical phonons and the photo-created electron-hole pair in the QD.

3 Calculation of excitonic polaron

The noninteracting exciton-phonon states are labelled $|n_e, n_h, N_{\mathbf{q}}\rangle$, where $|n\rangle = |s\rangle, |p^{\pm}\rangle$ are purely electronic levels. $|N_{\mathbf{q}}\rangle$ denotes the ensemble of the N LO-phonon states in the $\{\mathbf{q}\}$ modes. The Fröhlich Hamiltonian V_F couples states which differ by one phonon.

$$\langle n_e, n_h, 0|V_F|n'_e, n'_h, 1_{\mathbf{q}}\rangle = \frac{A_F}{q}\left[\delta_{n_h n'_h} v_{n_e n'_e}(\mathbf{q}) - \delta_{n_e n'_e} v_{n_h n'_h}(\mathbf{q})\right] \tag{1}$$

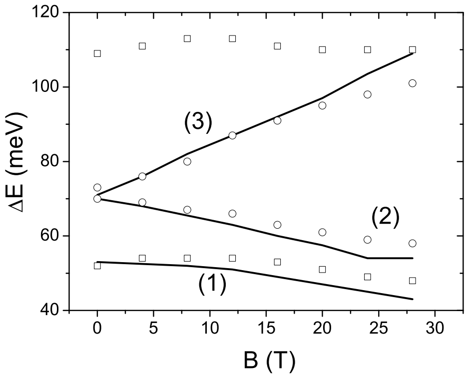

Fig. 2 Magnetic field dispersion of PLE resonances in sample A3 in open figures with the calculated energy transitions in solid lines.

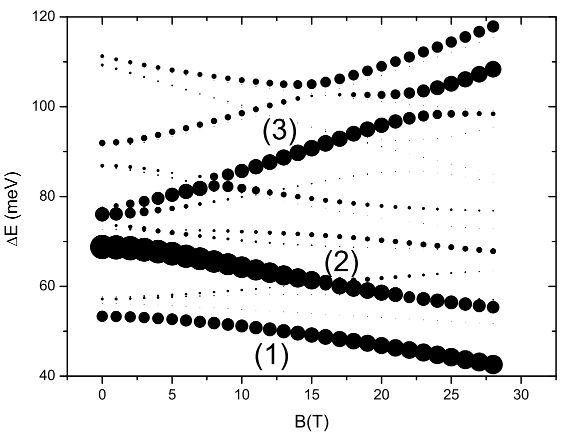

Fig. 3 Calculated excitonic polaron energies and intensities as a function of the magnetic field. The area of the circles are proportional to the oscillator strengths.

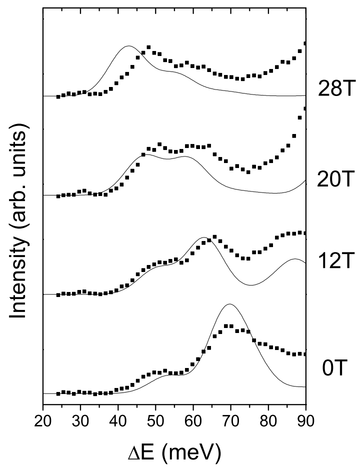

Fig. 4 Experimental (full squares) and calculated (solid lines) spectra for different magnetic fields. The experimental data was taken for E_{det}=1215 meV and for sample A3.

where $v_{nn'}(\mathbf{q}) = \langle n|e^{i\mathbf{q}\cdot\mathbf{r}}|n'\rangle$. The term A_F includes in its definition the Fröhlich constant which is taken as $\alpha = 0.11$, consistent with precedent polaron studies in QDs [1].

In order to calculate the excitonic polarons, we have used the dispersionless LO phonon approximation, which allows the very accurate calculation of polaron levels in QDs [1]. An LO phonon energy of $\hbar\omega$ = 36 meV has been used. In addition to Fröhlich coupling terms, coulomb interactions and in-plane anisotropy have been taken into account. The energy dispersions and oscillator strengths of the polaron states, presented in Fig. 3, were calculated using an in-plane effective mass for electrons m_e= 0.07m_o and for holes m_h= 0.22m_o, in agreement with FIR intraband magnetospectrocopy results. The other dots parameters were chosen in order to fit the experimental values of both the intraband s-p electronic (hole) transitions [47 meV (22 meV)] and the interband ground state energy (detection energy of 1215 meV). More details on polaron calculation can be found in reference [10]. The numerical diagonalization of the full Hamiltonian gives us the polaron energy transitions. The energy positions as a function of magnetic field are in good agreement with our data, as seen in Fig. 2 [11]. In particular, the Fröhlich coupling between the states $|p_e^-, p_h^+, 0\rangle$ and $|s_e, p_h^+, 1\rangle$ is responsible for the experimentally observed strong anticrossing. We calculate also the interband absorptions for an inhomogeneous ensemble of dots. The solid lines in Fig. 4 represent the calculated absorption spectra at different magnetic fields. We compare our calculated interband absorption spectra with the PLE experimental data. The full squares are data points taken for sample A3. The evolution of the oscillator strengths of the absorptions with the magnetic field is very well described by our model. We are able to predict the exchange of oscillator strength observed in our results demonstrating the validity of our analysis and the existence of excitonic polarons. Similar agreement is found for results obtained for sample A1 and A2.

4 Conclusion We have investigated the interband transitions in several ensembles of self-assembled InAs/GaAs QDs by using PLE spectroscopy. The magnetic field dependence of the interband transitions allows their unambiguous assignment. When two exciton-LO phonon states have close enough energies with phonon occupations which differ by one, a large anticrossing is theoretically predicted. Such a situation is experimentally induced in our samples by the applied magnetic field for the two interband transitions ($|p_e^-, p_h^+, 0\rangle, |s_e, p_h^+, 1\rangle$) and a strong anticrossing is actually observed in all the investigated samples. We have calculated the coupling between the mixed exciton-LO phonon states using the Fröhlich Hamiltonian and we have determined the energies and oscillator strengths of the interband transitions. Our model accounts well for the experimental data, evidencing that the excitons and LO-phonons are in a strong coupling regime in QDs and the interband transitions occur between excitonic polaron states. Finally, we believe that the existence of excitonic polarons could present important consequences for the energy relaxation in excited QDs and for the coherence decay times of the fundamental optical transitions.

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